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(INVESTIGATION OF ZEOLITE MEMBRANES FOR FUEL CELLS)

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1.0 INTRODUCTION AND SUMMARY

15875 *Author*

Investigation of the application of inorganic membranes in hydrogen-oxygen fuel cells has continued. Additional zirconium oxide-phosphoric acid-"Zeolon H" compositions have been prepared, processed, and tested for modulus of rupture and electrical conductivity.

A standard method for preparing inorganic membranes by either ball milling or high speed mixing is described in detail.

The addition of zirconia fibers to zirconium phosphate-"Zeolon H" membrane compositions resulted in a substantial increase in strength. Modulus of rupture values as high as 2751 psi were obtained compared to 1063 psi for the material without fibers. Increased transverse strength was also obtained through the use of more reactive grades of zirconia.

An experimental design has been established to permit statistical analysis of the significant variables in membrane preparation.

A number of preliminary fuel cell runs were made in order to improve fuel cell design for the evaluation of membrane characteristics. Favorable preliminary results were obtained, and typical polarization curves are shown at temperatures from 80° to 120°C. *Author*

2.0 EXPERIMENTAL WORK AND DISCUSSION

2.1 Membrane Preparation

2.1.1 Standard Method for the Preparation of Inorganic Membrane Materials

In order to produce satisfactory inorganic membranes for fuel cell application, it was necessary to rigidly standardize all the materials and procedures used in the preparation of the membranes. This accomplished, the application of adequate quality control practice to all phases of membrane preparation was carried out in order to produce membranes of uniform physical, electrical, and chemical characteristics. The following procedures have been established:

a. Ball Milling Membrane Compositions

Clean grinding jar, cover, gasket, and grinding balls by washing thoroughly. Place balls in grinding jar, cover with water, secure cover, and grind for 15 min. Empty mill and rinse with clean water. Repeat twice. Place grinding jar, cover and balls in drying oven at 120°C until completely dry.

Place the level of the grinding balls just above the midpoint of the jar (55% of the jar volume). For small grinding jars (1 gal and under), use 1-in. and smaller grinding balls. A gradation of sizes is preferable.

Weigh raw materials carefully and accurately so as to avoid spillage or contamination. Keep the grinding jar covered at all times during weighing in order to avoid pickup of airborne dust. Add dry materials to the grinding jar first, liquids last.

Milling time must be exact to a total variation of 5 min.

After grinding, empty the milled material and balls through an 8 mesh screen into a clean container. Screen this slurry through an 80 mesh screen. Place the screened slurry in suitable containers for drying.

Wash, dry, and return all equipment to storage.

b. Mixing Membrane Compositions

Clean mixing jar, agitator, and cover by washing thoroughly. Run with clean water for 5 to 10 min, empty, and rinse with clean water. Dry completely.

Add liquid ingredients to mixer first, then dry materials.

Put cover in place, start mixer, and run at low speed until dry materials are incorporated into liquid phase. Run at high speed for desired period of time.

Screen slurry through an 80 mesh screen and place in suitable container for drying. Wash and dry all equipment and return to storage.

2.1.2 Alternate Method for the Preparation of Inorganic Membrane Materials by Mixing

As all of the materials presently used in the preparation of the inorganic membranes are very finely divided (less than 10 microns), grinding in a ball mill is not necessary to reduce particle size. Accordingly, the results of mixing the raw materials in a high speed blender were compared with ball mill preparation. A standard 1 zirconia: 1 phosphoric acid: 1 "Zeolon H" mixture was made by mixing for 5 min in a Waring Laboratory Blender. After mixing, the slurry was dried for 15 hr at 160°C, granulated to -32 +80 mesh, and pressed into test membranes for sintering. Modulus of rupture specimens were cut from sample membranes and tested for strength. It was found that the strength of membranes prepared by this mixing technique was as satisfactory as those prepared by ball milling.

Preparation of inorganic membrane materials by mixing not only greatly reduces preparation time but also offers the advantage of being able to introduce materials into the composition without significantly altering their physical form. Fibers, for example, can be added to the mixture by this means, whereas ball milling might largely destroy the fibrous structure due to the grinding action of the media.

When particle size reduction is desired, a combination of the two preparation techniques may be used, especially when fibers of similar materials are to be added to the composition. In this case, ball milling would be used to grind the raw materials to the desired degree of fineness, and then the fibers or other additives would be introduced into the composition by high speed mixing.

2.1.3 Preparation of Membrane Materials for Compacting

The dried membrane material is crushed with a porcelain mortar and pestle and passed through a 32 mesh screen. The fines are removed with an 80 mesh screen, resulting in a -32 mesh, +80 mesh product.

The pressing material is stored in a sealed jar to prevent pickup (or loss) of moisture due to excessive exposure to the atmosphere.

2.1.4 Compacting Membrane Materials

The pressing mold is cleaned thoroughly and lubricated with a thin film of kerosene. Both punches are checked for fit and freedom of movement in the die cavity.

The bottom punch is then inserted into the cavity and blocked at the proper level to provide the desired die fill. The die cavity is "over-filled" with an excess of material, and a smooth, uniform final die fill is obtained by carefully scraping off the excess material with a spatula. Care is taken to avoid removing any material from the die cavity. The top punch is then inserted into the die cavity and checked to ascertain that it fits and turns freely. The blocking which supports the lower punch, is removed, a rubber gasket ring is placed over the lower punch, and the mold assembly is placed in the press. The mold assembly is pressed lightly (>5 tons load) to exclude air, pressure is released, and then the desired load is applied. Pressure is released slowly, and the mold assembly is removed from the press. The gasket ring is then removed, and the upper punch is pressed through the mold assembly to eject the pressed compact.

The lower punch (now in top position) is removed, followed by removal of the compacted membrane. As the membrane is very soft and fragile at this stage, extreme care must be used to avoid cracking, breakage, or distortion. The membrane can usually be floated from the punch with a gentle jet of compressed air and transferred to a smooth, flat firing plate by means of a wide spatula.

After compacting, the membranes are stored in a drying oven (110°C) or a dessicator until they can be loaded into the furnace for sintering.

2.1.5 Sintering the Membranes

The membranes can be sintered by loading them on smooth, flat refractory surfaces and placing them in the electric sintering furnace. Refractory setters commonly used for firing ceramic wall tile have been found to be highly satisfactory for this purpose. They are flat and smooth, nine membranes can be loaded on each setter, and the setters can be stacked four high in the present sintering furnace so that as many as 36 membranes can be sintered

at one time. The setters also provide a constant thermal load (mass) for the furnace, which increases the uniformity of sintering within a given run and from load to load.

After sintering at the desired temperature and time, the furnace is allowed to cool to 110°C, and the refractory setting and sintered membranes are removed. The membranes are then stored in sealed glass jars to avoid moisture pickup from the atmosphere.

2.2 Membrane Composition Studies

2.2.1 Addition of Zirconia Fibers to Zirconium Phosphate-"Zeolon H" Membranes

Favorable results have been obtained through the addition of fiber to ceramics in an Astropower proprietary program. Therefore, it was deemed desirable to determine the effect of fiber additions in this project. Refractory fibers commercially available at the present time include calcia-stabilized zirconia, zirconium silicate, calcia-stabilized zirconia containing 9 to 10% silica, calcia-stabilized zirconia containing 10 to 15% neodymia, alumino-silicate, silica, glass, and acid-washed asbestos. Calcia-stabilized zirconia was selected for evaluation as an additive to zirconium phosphate-"Zeolon H" membranes because of its refractiveness, strength, and chemical compatibility with the other membrane ingredients. It was felt that the fibers should increase membrane strength both by virtue of their physical form and chemical reaction with the phosphoric acid present in the mixture. The physical properties of calcia-stabilized zirconia are shown in Table I.

Test membranes were prepared from a 1 zirconia: 1 phosphoric acid: 1 "Zeolon H" composition which was ball milled for 18 hr. After grinding, 5% by weight of TAM XPL-100-2 zirconia fibers were added to the slurry and mixed in a Waring Blendor for 5 min. This material was then dried, granulated, pressed, and sintered. Modulus of rupture samples were cut from sintered membranes and tested for transverse strength. Also, conductivities were measured, and membranes were tested in the experimental fuel cell.

The data obtained show that the addition of 5% calcia-stabilized zirconia fibers to a 1 zirconia: 1 phosphoric acid: 1 "Zeolon H" membrane composition increased the transverse strength from 1063 psi to 1910 psi when sintered at 300°C. Sintering at 500°C resulted in a modulus of rupture of 2751 psi for the composition containing 5% ZrO₂ fiber, compared to 2381 psi for the 1 ZrO₂: 1 H₃PO₄: 1 "Zeolon H" composition without fiber addition.

Transverse strengths are shown on Figure 1 and in Table II. Composition and processing details are shown in Table III.

Favorable results were obtained when the fiber-containing membrane was operated in the test fuel cell. As a result of these findings, the effects of adding zirconia fibers to zirconium phosphate-"Zeolon H" fuel cell membranes will be investigated further.

2.2.2 Evaluation of Zirconium Oxides from Different Sources

Investigations conducted in Astropower proprietary programs involving the reactivity of zirconium oxides obtained from different sources with phosphoric acid suggested that further improvement in membrane strength could result from the use of more reactive zirconias. A sample of C. P. Zirconia was obtained from Titanium-Zirconium Co., Flemington, N.J., which was believed to have been calcined to a lower temperature than the TAM C.P. zirconia being used and would be more reactive. Sample membranes were prepared from a standard 1/1/1 zirconia/phosphoric acid/"Zeolon H" mixture using this material as the zirconia constituent. The increased reactivity of this zirconia was readily apparent during drying of the material after mixing, and from the noticeable amount of heat which was generated when it was mixed with phosphoric acid. The strength of the sintered membranes was also excellent (2941 ± 210 psi) compared to the same composition when TAM C.P. zirconia was used (1063 ± 90 psi). As a result of these favorable preliminary findings, additional lots of zirconia calcined to lower temperatures have been ordered for further evaluation. Also, samples of calcia-stabilized zirconia are being obtained, as it is reasonable to believe that these materials should also develop stronger bonds with phosphoric acid than the C.P. grades of zirconia.

2.3 Conductivity Measurements

The conductivities of three different types of zirconium oxide-phosphoric acid-"Zeolon H" membranes were determined at a number of fixed temperatures in the vicinity of 100°C . The effect of temperature as well as of relative humidity was measured. The apparatus employed for these measurements was previously described in the third quarterly report (108-Q3). In this arrangement, the alternating current resistance of membranes is measured by a bridge method. In order to assess the reproducibility of membrane preparation, the conductivities of a number of identical membranes were determined. The resistivities presented represent average values; the error is indicated in terms of standard deviation.

The resistivities of two new membrane compositions (Nos. 191-017-1 and 191-016), consisting of ZrO_2 , H_3PO_4 , and "Zeolon H" (1:2:3/2 and 2:1:2 parts by weight) are shown in Tables IV and VI as a function of both temperature and relative humidity. As a standard for comparison, the usual highly conducting membrane composition consisting of ZrO_2 , H_3PO_4 , and "Zeolon H" (1:1:1) was prepared under the same conditions and is shown in Table V. As illustrated by Figures 2 and 3, the new membrane composition consisting of ZrO_2 , H_3PO_4 , and "Zeolon H" (1:2:3/2) exhibits a somewhat lower value of resistivity than the highly conducting composition (1:1:1). For example, at 60% relative humidity and 90°C , a resistivity of 29 ohm-cm is exhibited by 1:2:3/2, while 35 ohm-cm is shown by 1:1:1. A further examination of these figures reveals that both membrane compositions show increasing conductivity with an increase of either the relative humidity or the temperature, as would be expected for ionic conduction; however, the relative effect of temperature is quite small.

The remaining new membrane composition, Membrane No. 191-016 formed from ZrO_2 , H_3PO_4 , and "Zeolon H" (2:1:2) is considerably less conducting than the 1:1:1 membrane composition, probably due to the excess unreacted ZrO_2

caused by using a low H_3PO_4 ratio. The resistivity of this membrane (at 90°C and 70% relative humidity) is 92 ohm-cm, compared to 35 for the 1:1:1 membrane.

In Figure 4 the resistivities of all three membrane compositions are compared at 110°C as a function of relative humidity. Again, this plot shows the same order of conductivities as deduced at lower temperatures. The plot shows clearly that even at 110°C the 2:1:2 mixture is considerably less conducting than either the 1:2:3/2 or 1:1:1, which exhibit quite similar conductivities.

2.4 Fuel Cell Operation

A single-cell laboratory fuel cell has been constructed for use in the evaluation of the more promising membranes. A cross section of this fuel cell as well as a description of its construction is presented in the previous monthly report (108-M2). Photographs of the cell assembled and disassembled are shown in Figures 5 and 6.

The entire system employed in the control and measurement of each variable such as temperature, gas flow, current flow, and voltage is shown in the block diagram of the preceding monthly report (108-M2). In this arrangement, the fuel cell is immersed in a fluidized sand bath which is maintained at the desired temperature by means of a solid state proportional temperature controller that allows temperature control to within less than 1°C . Both hydrogen and oxygen gases are measured by flow meters before entering the fuel cell. The flow rates can be carefully adjusted by low pressure regulators preceding the flow meters. For determination of current-potential curves, the fuel cell is connected in series with a Western milliammeter and a low resistance transistorized power supply. This arrangement assures constant current operation, allowing the fuel cell to determine the direction of current while the power supply compensates for minor variations in the output voltage of the cell. Finally, the voltage output of the fuel cell is measured by a high impedance Triplet electronic voltmeter.

A few preliminary experiments have been carried to test and refine the fuel cell apparatus. In these experiments, the membrane was introduced into the fuel cell, and silicone rubber was placed around the perimeter of the membrane to achieve a gas seal. The platinum black catalyst was applied mechanically upon each side of the membrane as well as upon a 50 mesh platinum screen held in contact with the membrane by spring-loaded pusher plates. The fuel cell was assembled and introduced into the fluidized sand bath maintained at 25°C . The membrane in the fuel cell was hydrated at room temperature by short circuiting the electrodes while hydrogen and oxygen gases flowed through the compartments on each side of the membrane. After appreciable hydration of the membrane took place, significant open circuit potentials were observed.

After pretreatment of the membrane, the effect of temperature and of the rate of flow of gases (which determine the water balance within the membrane) upon the current-potential curves, as well as upon the open circuit potential, were investigated.

2.5 Results

Employing the procedure described above, Membrane No. 191-002, consisting of ZrO_2 , H_3PO_4 , and "Zeolon H" (1:1:1 parts by weight with addition of 5% ZrO_2 fibers) was tested in the fuel cell assembly. The potential-current data at a number of temperatures from 80°C to 120°C are shown in Table VII and illustrated in Figure 7. Inspection of this table reveals that a fairly satisfactory open circuit potential of 0.96 v is observed at 80°C and gradually decreases with increasing temperatures to a value of 0.84 v at 120°C . As shown by Figure 7, the current output at a given voltage level improved with increasing temperatures up to 90°C , whereupon, as the temperature was further increased to 100, 110, and 120°C , the performance of the fuel cell was lowered. At this optimum temperature of 90°C , a current density of 31 ma/cm^2 was sustained at 0.5 v output. From the slopes of the potential-current curves, the apparent membrane resistance is calculated as shown in Table VIII. Inspection of this table reveals that the minimum membrane resistance of 1 ohm is obtained at 90°C , with a maximum value of 3 ohms at 120°C . If these resistance values are expressed in terms of membrane resistivity, a minimum value of 170 ohm-cm is obtained at 90°C , while the maximum of 509 ohm-cm occurs at 120°C . These unusually large values for resistivity (about tenfold larger than may be expected from conductivity measurements) may be a result of:

- a. Inadequate coverage of the membrane by platinum black catalyst, leaving areas of the membrane without catalyst
- b. A contact resistance between the platinum black and the membrane
- c. Electrode flooding so that large areas of the electrode surface are rendered ineffective for electrode reaction

The above fuel cell tests were preliminary in nature and are not a satisfactory criterion of membrane characteristics. As the method of contacting the membrane with platinum black catalyst is improved and other difficulties are eliminated, a fairly good evaluation of the membrane characteristics may be anticipated. A number of different approaches for improving the application of platinum to the membrane surface are currently being explored and evaluated.

2.6 Statistical Design

In order to determine the optimum procedure for fabricating inorganic fuel cell membranes, an experimental design has been established. Preliminary experiments have been carried to determine limits for drying time and temperatures. The experimental plan is shown in Table IX. The experimental work necessary to provide data for statistical analysis is under way, and preliminary results should be available for the next report.

3.0 PERSONNEL CHANGES

Mr. A. D. Kelmers has left Astropower, Inc. Dr. Duncan W. Cleaves and Mr. W. H. Graves, Jr. are now associated with the Electrochemistry Department.

Dr. Cleaves has extensive experience in ion exchange technology and has carried out research in both the inorganic and organic fields. His background includes 14 years of industrial chemical experience, and 6 years of college teaching. He assisted in or directed a variety of laboratory and pilot scale problems in chemical process research, including organic esterification, ion-exchange separation of amino acids, electrodeposition of copper and the extractive metallurgy of titanium. He adapted the Kroll process to the production of high purity titanium. He has designed and built glass vacuum systems for the analysis of metals by hot-extraction and vacuum fusion techniques.

Mr. Graves has broad experience in research and engineering in the fields of fuel cells, petroleum, medicine, textiles, and specialized analytical instrumentation. He has also developed high temperature-high pressure research equipment, and he is the inventor of devices for use in radioactive gas sampling and handling.

TABLE I
PHYSICAL PROPERTIES OF CALCIA-STABILIZED
ZIRCONIA FIBERS*

Stabilized zirconia, 4 to 6% calcia

Fiber length	1-3 in.
Fiber diameter	0.0001-0.0003 in.
Cross section	Circular
Grain size	0.1 micron
Specific gravity	4.5-5.2
Melting point	2550°C
Tensile strength	35,000-150,000 psi
Modulus of elasticity	$30-50 \times 10^6$ psi
Surface area	200 m ² /g

* H. I. Thompson Fiber Glass Co. Technical Bulletin
 No. 1-1B. ✓

TABLE II
DRY TRANSVERSE STRENGTH OF SINTERED MEMBRANES

<u>Batch No.</u>	<u>ZrO₂</u>	<u>Phosphoric Acid</u>	<u>"Zeolon H"</u>	<u>Modulus of Rupture</u>
191-019	1	1	0	4135 \pm 24 psi
191-020	1	1/2	1	2405 \pm 70 psi
191-007	1	1	1	1063 \pm 90 psi
191-002	1	1	1	1910 \pm 210 psi
191-042	1	1	1	2751 \pm 59 psi
036-029	1	1	1	2381 \pm 115 psi

TABLE III
COMPOSITION AND PROCESSING METHODS USED IN PREPARING
FUEL CELL MEMBRANES

<u>Membrane No.</u>	<u>Composition</u>			<u>Ball Milling</u>	<u>Drying</u>		<u>Pressing Pressure</u>	<u>Sintering</u>	
	<u>ZrO₂</u>	<u>Phos Acid</u>	<u>"Zeolon H"</u>		<u>Temp</u>	<u>Time</u>		<u>Temp</u>	<u>Time</u>
191-019	1	1	0	18 hr	160°C	15 hr	5 tons	300°C	24 hr
191-020	2	1	2	18 hr	160°C	15 hr	5 tons	300°C	24 hr
191-007	1	1	1	18 hr	160°C	15 hr	15 tons	300°C	24 hr
191-002 ⁽³⁾	1	1	1	18 hr	160°C	15 hr	15 tons	300°C	24 hr
191-042 ⁽³⁾	1	1	1	(1)	160°C	15 hr	15 tons	500°C	24 hr
036-029 ⁽²⁾	1	1	1	(1)	160°C	15 hr	15 tons	500°C	24 hr

(1) Not ball milled - mixed 15 min in Waring blender

(2) C. P. zirconia from Titanium-Zirconium Co., Flemington, N. J.

(3) 5% TAM ZrO₂ fibers; XPL-108-2 added after preparation

TABLE IV
RESISTIVITIES AT THREE TEMPERATURES
(MEMBRANE NO. 191-017-1)

Composition: ZrO_2 , H_3PO_4 , "Zeolon H" ($1:2:\frac{3}{2}$ parts by weight)

<u>Relative Humidity</u> <u>(%)</u>	<u>Resistivity</u> <u>(Ohm-cm)</u>	<u>Number of Membranes Employed</u> <u>in Obtaining Standard Deviations</u>
<u>Temperature 69°C</u>		
69	$(3.12 \pm 0.18) \times 10^1$	3
56	$(3.31 \pm 0.22) \times 10^1$	3
42	$(3.48 \pm 0.26) \times 10^1$	3
25	$(4.44 \pm 0.44) \times 10^1$	3
15	$(7.72 \pm 0.95) \times 10^1$	3
0	$(8.94 \pm 0.80) \times 10^2$	3
<u>Temperature 90°C</u>		
70	$(2.88 \pm 0.24) \times 10^1$	3
49	$(2.94 \pm 0.29) \times 10^1$	3
28	$(5.04 \pm 0.63) \times 10^1$	3
20	$(6.11 \pm 0.83) \times 10^1$	3
11	$(8.81 \pm 1.27) \times 10^1$	3
0	$(2.47 \pm 0.57) \times 10^3$	3
<u>Temperature 110°C</u>		
57	$(2.47 \pm 0.15) \times 10^1$	3
32	$(4.12 \pm 0.29) \times 10^1$	3
14	$(1.05 \pm 0.09) \times 10^2$	3
9	$(1.25 \pm 0.12) \times 10^2$	3
7	$(1.33 \pm 0.14) \times 10^2$	3
5	$(1.65 \pm 0.67) \times 10^2$	3
0	$(2.90 \pm 0.38) \times 10^2$	3

TABLE V
RESISTIVITIES AT THREE TEMPERATURES
(MEMBRANE NO. 191-007-1)

Composition: ZrO_2 , H_3PO_4 , "Zeolon H" (1:1:1 parts by weight)

<u>Relative Humidity</u> <u>(%)</u>	<u>Resistivity</u> <u>(Ohm-cm)</u>	<u>Number of Membranes Employed</u> <u>in Obtaining Standard Deviations</u>
<u>Temperature 69°C</u>		
69	$(3.80 \pm 0.07) \times 10^1$	3
56	$(3.97 \pm 0.05) \times 10^1$	3
42	$(4.16 \pm 0.06) \times 10^1$	3
25	$(6.57 \pm 0.76) \times 10^1$	3
15	$(1.06 \pm 0.24) \times 10^2$	3
0	$(1.46 \pm 0.45) \times 10^3$	3
<u>Temperature 90°C</u>		
70	$(3.48 \pm 0.06) \times 10^1$	3
49	$(3.65 \pm 0.16) \times 10^1$	3
28	$(6.86 \pm 1.39) \times 10^1$	3
20	$(8.01 \pm 1.58) \times 10^1$	3
11	$(1.12 \pm 0.23) \times 10^2$	3
0	$(2.57 \pm 0.67) \times 10^3$	3
<u>Temperature 110°C</u>		
57	$(3.22 \pm 0.12) \times 10^1$	3
32	$(6.09 \pm 1.54) \times 10^1$	3
14	$(1.47 \pm 0.34) \times 10^2$	3
9	$(1.84 \pm 0.42) \times 10^2$	3
7	$(1.97 \pm 0.45) \times 10^2$	3
5	$(2.52 \pm 0.59) \times 10^2$	3
0	$(4.76 \pm 1.40) \times 10^2$	3

TABLE VI
RESISTIVITIES AT THREE TEMPERATURES
(MEMBRANE NO. 191-016)

Composition: ZrO_2 , H_3PO_4 , "Zeolon H" (2:1:2 parts by weight)

<u>Relative Humidity</u> <u>(%)</u>	<u>Resistivity</u> <u>(Ohm-cm)</u>	<u>Number of Membranes Employed</u> <u>in Obtaining Standard Deviations</u>
<u>Temperature 69°C</u>		
69	$(7.61 \pm 1.13) \times 10^1$	2
56	$(1.13 \pm 0.20) \times 10^2$	2
42	$(1.59 \pm 0.31) \times 10^2$	2
25	$(2.91 \pm 0.18) \times 10^2$	2
15	$(6.16 \pm 1.51) \times 10^2$	2
0	$(2.26 \pm 0.82) \times 10^4$	2
<u>Temperature 90°C</u>		
70	$(0.92 \pm 0.16) \times 10^2$	2
49	$(1.62 \pm 0.28) \times 10^2$	2
28	$(3.81 \pm 0.85) \times 10^2$	2
20	$(4.66 \pm 1.09) \times 10^2$	2
11	$(7.17 \pm 1.85) \times 10^2$	2
0	$(2.68 \pm 0.92) \times 10^4$	2
<u>Temperature 110°C</u>		
57	$(0.92 \pm 0.14) \times 10^2$	2
32	$(2.43 \pm 0.44) \times 10^2$	2
14	$(5.98 \pm 1.20) \times 10^2$	2
9	$(7.34 \pm 1.41) \times 10^2$	2
7	$(8.13 \pm 1.65) \times 10^2$	2
5	$(9.95 \pm 1.98) \times 10^2$	2
0	$(4.35 \pm 0.42) \times 10^3$	2

TABLE VII
EFFECT OF TEMPERATURE UPON CURRENT-POTENTIAL
CHARACTERISTICS OF A HYDROGEN-OXYGEN MEMBRANE
FUEL CELL

Membrane No. 191-002

Composition: ZrO_2 , H_3PO_4 , "Zeolon H" (1:1:1 parts by weight with addition of 5% ZrO_2 fibers)

Catalyst: Platinum black

Method of catalyst contact with membrane: platinum black applied mechanically to 50 mesh platinum screen in contact with membrane

Apparent area of platinum screen = 13.4 cm^2

<u>Temperature</u> <u>80°C</u>		<u>Temperature</u> <u>90°C</u>		<u>Temperature</u> <u>100°C</u>		<u>Temperature</u> <u>110°C</u>		<u>Temperature</u> <u>120°C</u>	
V	I	V	I	V	I	V	I	V	I
(volts)	(ma)	(volts)	(ma)	(volts)	(ma)	(volts)	(ma)	(volts)	(ma)
0.96	0	0.90	0	0.86	0	0.87	0	0.84	0
0.70	150	0.71	200	0.64	150	0.62	100	0.53	100
0.64	200	0.62	300	0.56	200	0.39	200	0.38	150
0.57	250	0.52	400	0.38	300	0.17	300	0.23	200
0.47	300	0.39	500	0.23	400	0.05	350	0.07	250
0.41	350	0.30	600	0.02	500				
0.35	400	0.21	700						
0.23	500	0.13	800						
0.11	600								

TABLE VIII
EVALUATION OF THE RESISTIVITY OF A MEMBRANE AT A
NUMBER OF DIFFERENT TEMPERATURES FROM
CURRENT-POTENTIAL DATA

Membrane No. 191-002

Apparent electrode area = 13.4 cm²

Thickness of membrane = 7.9 x 10⁻² cm

<u>Temperature</u> <u>(°C)</u>	<u>Resistance</u> <u>(Ohms)</u>	<u>Resistivity</u> <u>(Ohm-cm)</u>
80	1.4	238
90	1.0	170
100	1.8	305
110	2.2	373
120	3.0	509

TABLE IX
STATISTICAL DESIGN FOR DETERMINING OPTIMUM
PROCEDURES FOR FABRICATING MEMBRANES

<u>Variable</u> <u>Elements</u>	<u>Composition</u>			
X_1	30	30	30	Zirconia
	20	30	40	Phosphoric acid
	50	40	30	"Zeolon H"
	<u>Drying Time</u>			
X_2	20	40	60	Hr
	<u>Drying Temperature</u>			
X_3	120	140	160	°C

<u>Fixed</u> <u>Elements</u>	<u>Compaction Pressure</u>			
	15	15	15	Tons
	<u>Sintering Time</u>			
	24	24	24	Hr
	<u>Sintering Temperature</u>			
	500	500	500	°C

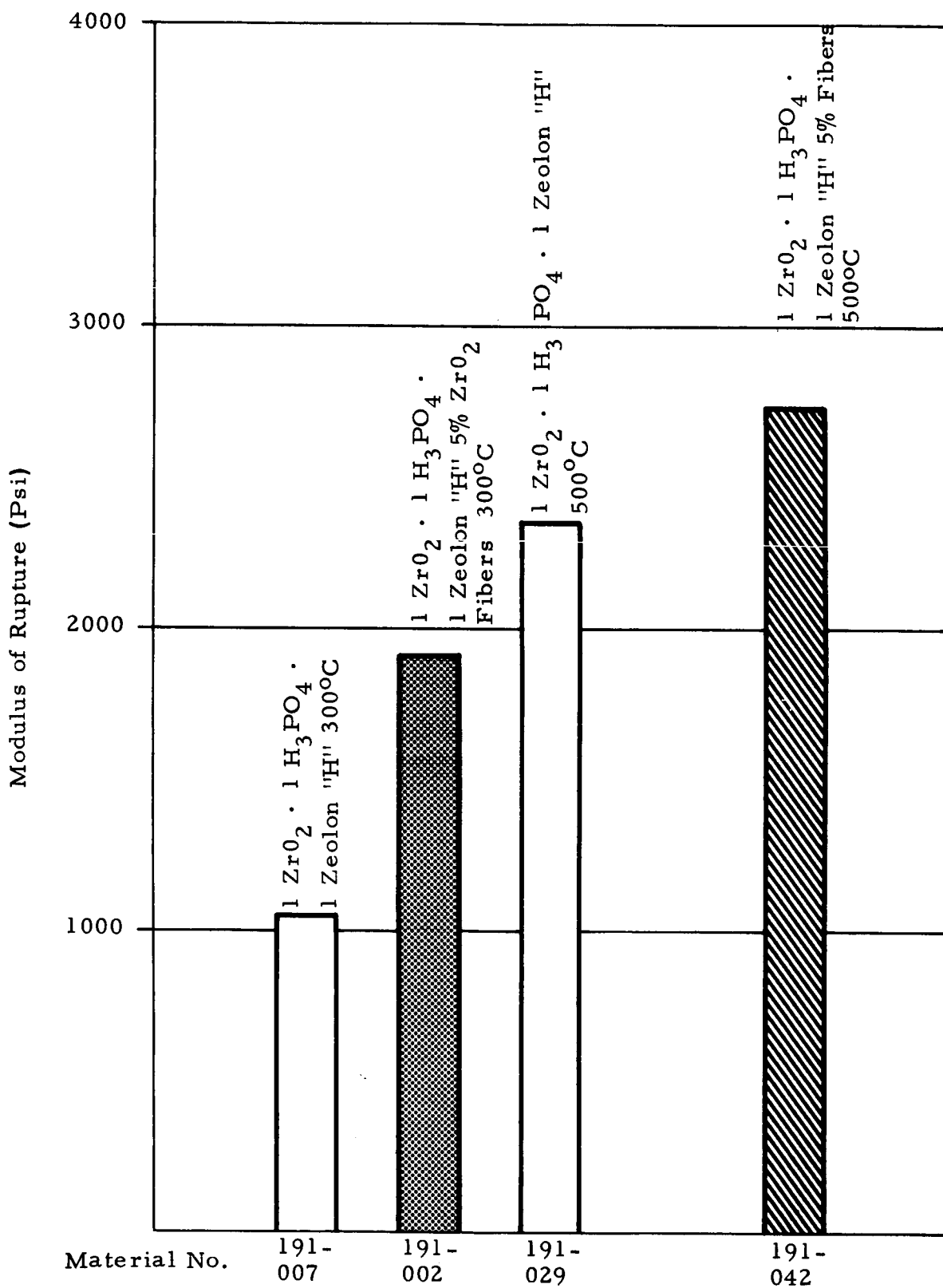


Figure 1. Effect of Zirconia Fiber Additions on the Transverse Strength of 1 $\text{ZrO}_2 \cdot \text{H}_3\text{PO}_4$ · 1 Zeolon "H" Membranes

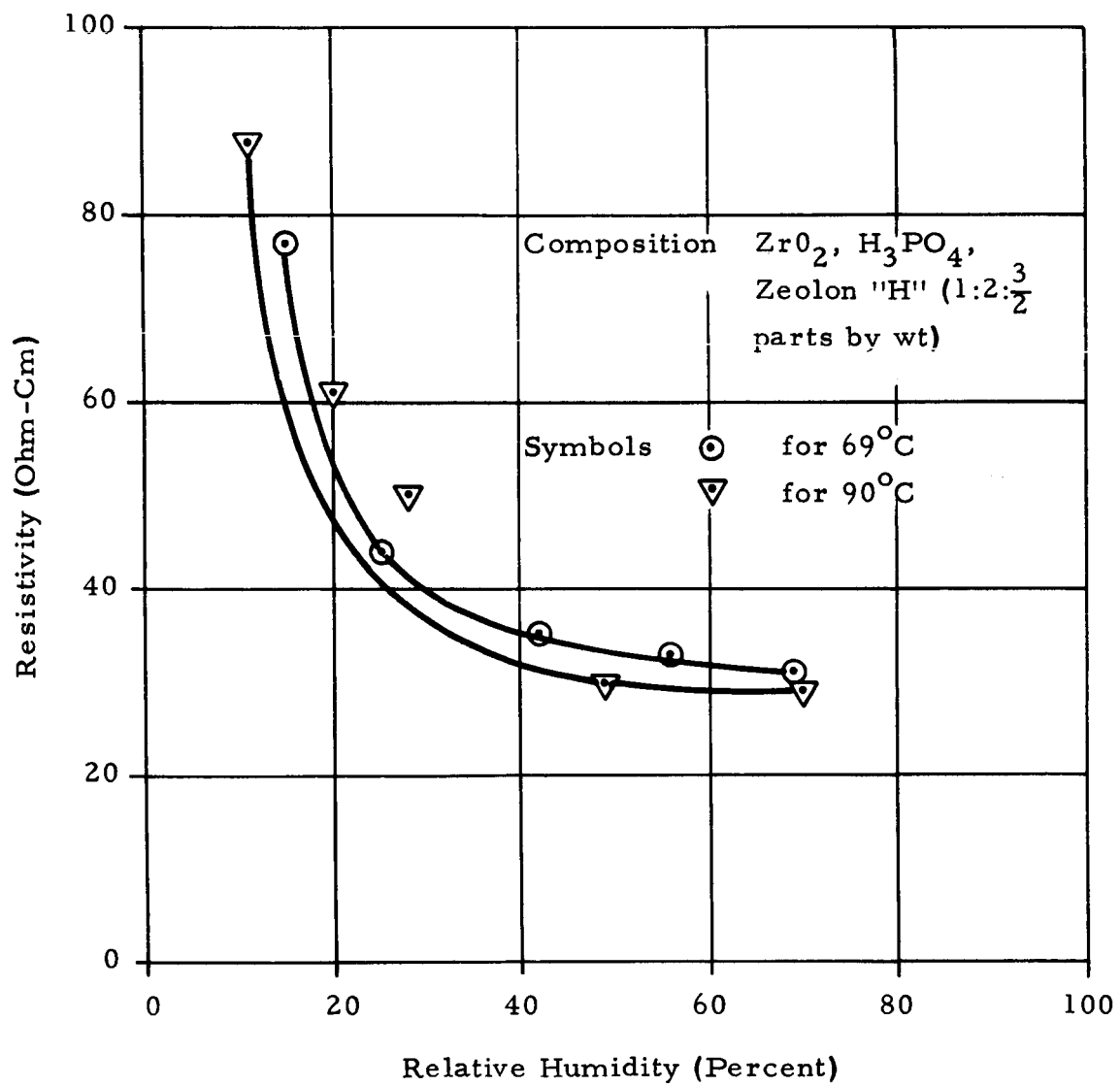


Figure 2. Effect of Temperature upon Resistivity for the Same Membrane Composition (Membrane No. 191-017-1)

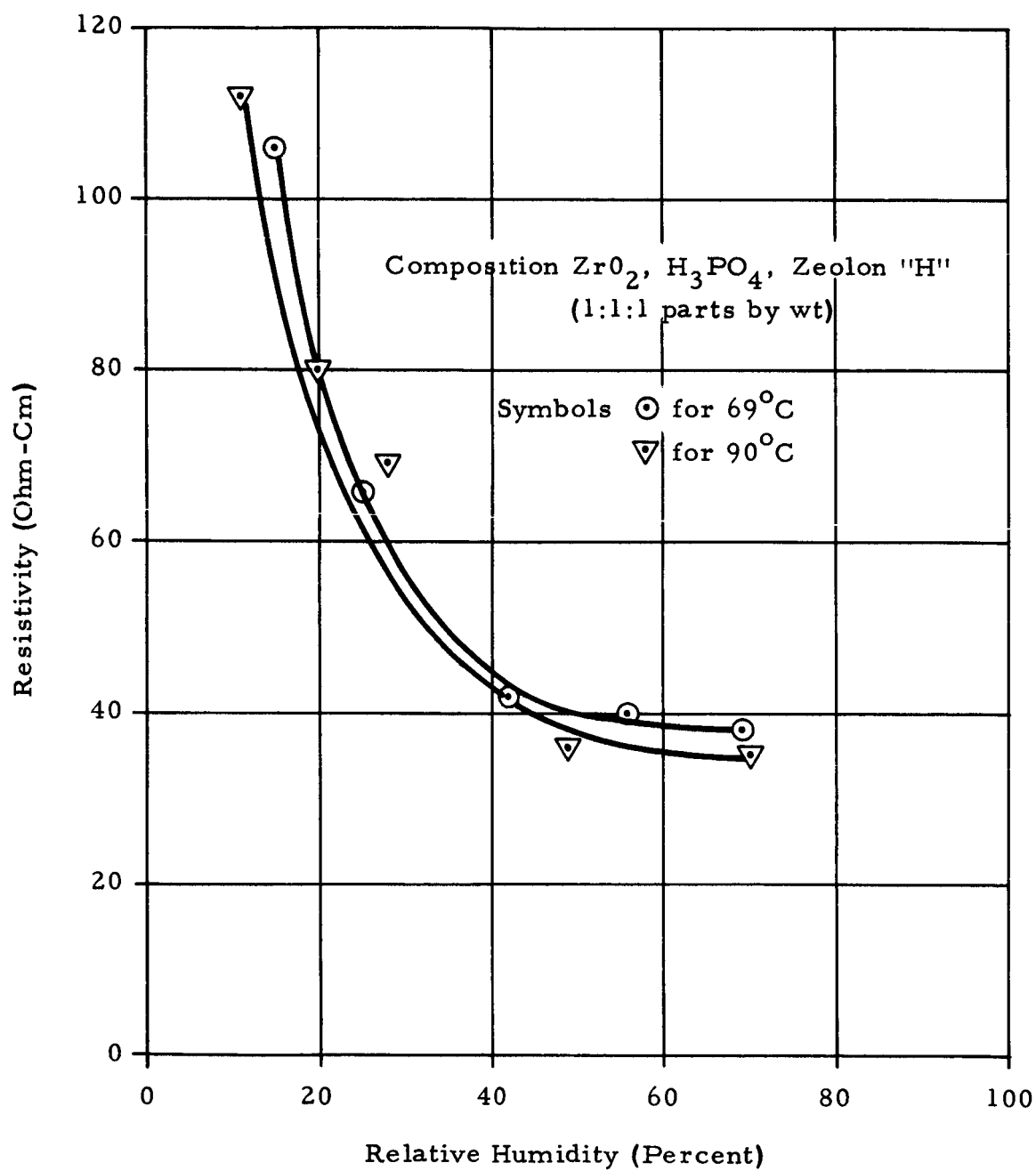


Figure 3. Effect of Temperature upon Resistivity for the Same Membrane Composition (Membrane No. 191-007-1)

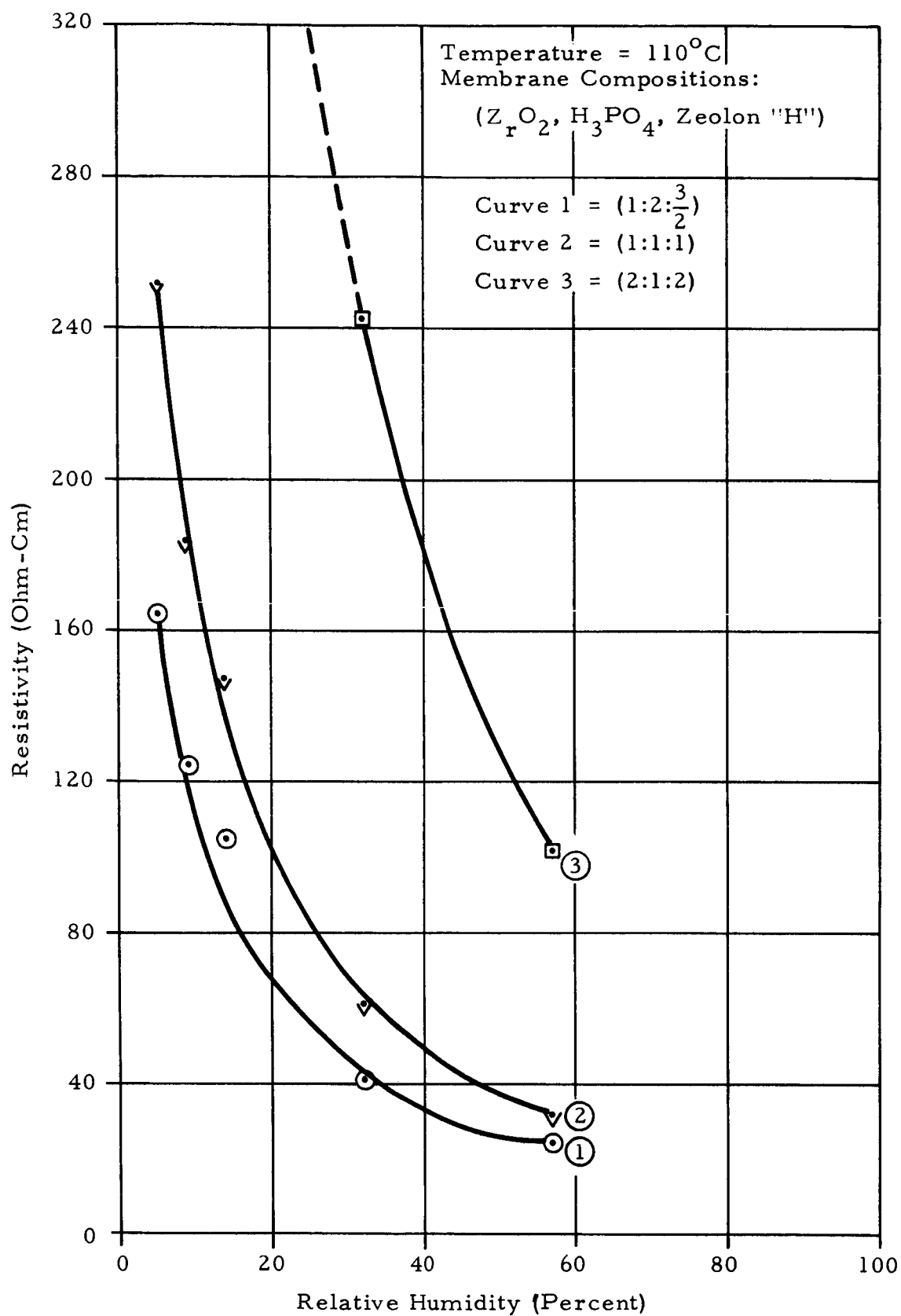


Figure 4. Comparison of Resistivities for Three Different Membrane Compositions

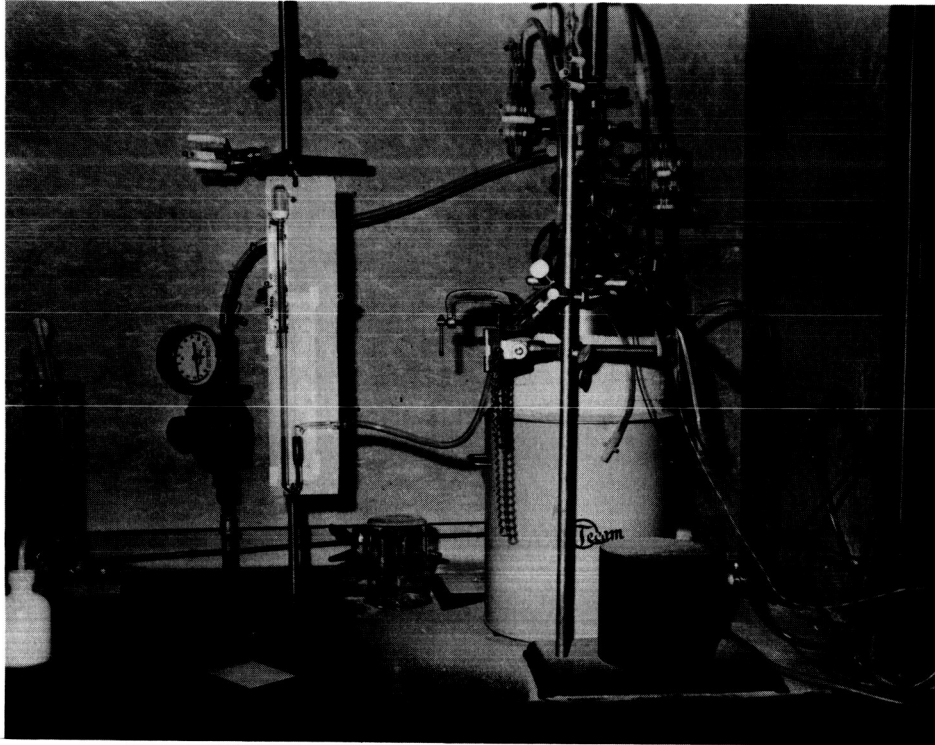


Figure 5. Assembled Fuel Cell

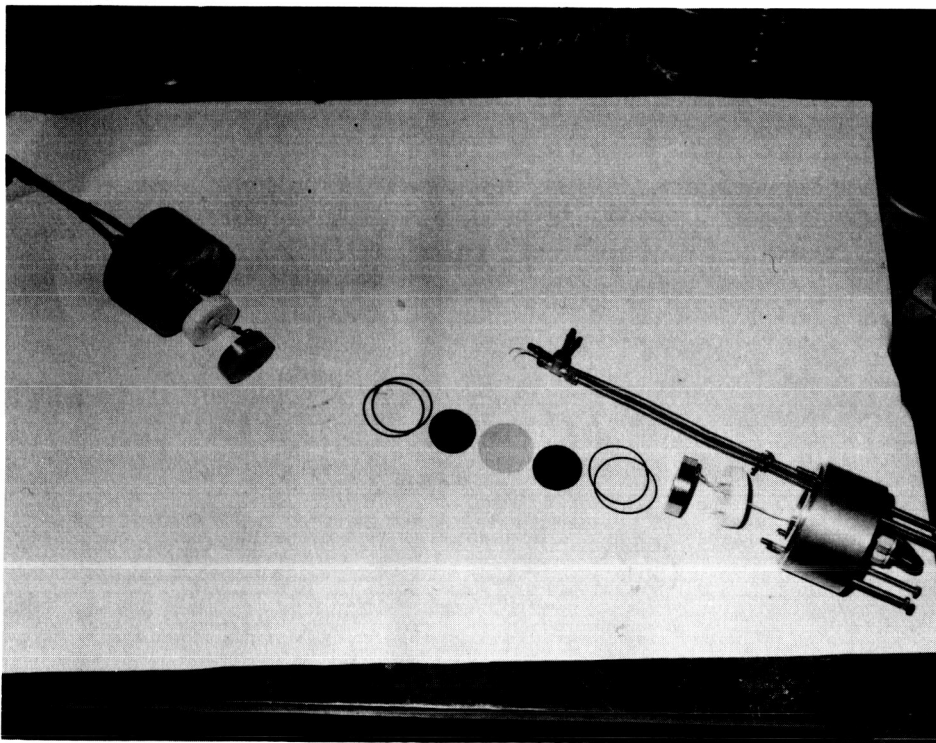


Figure 6. Disassembled Fuel Cell

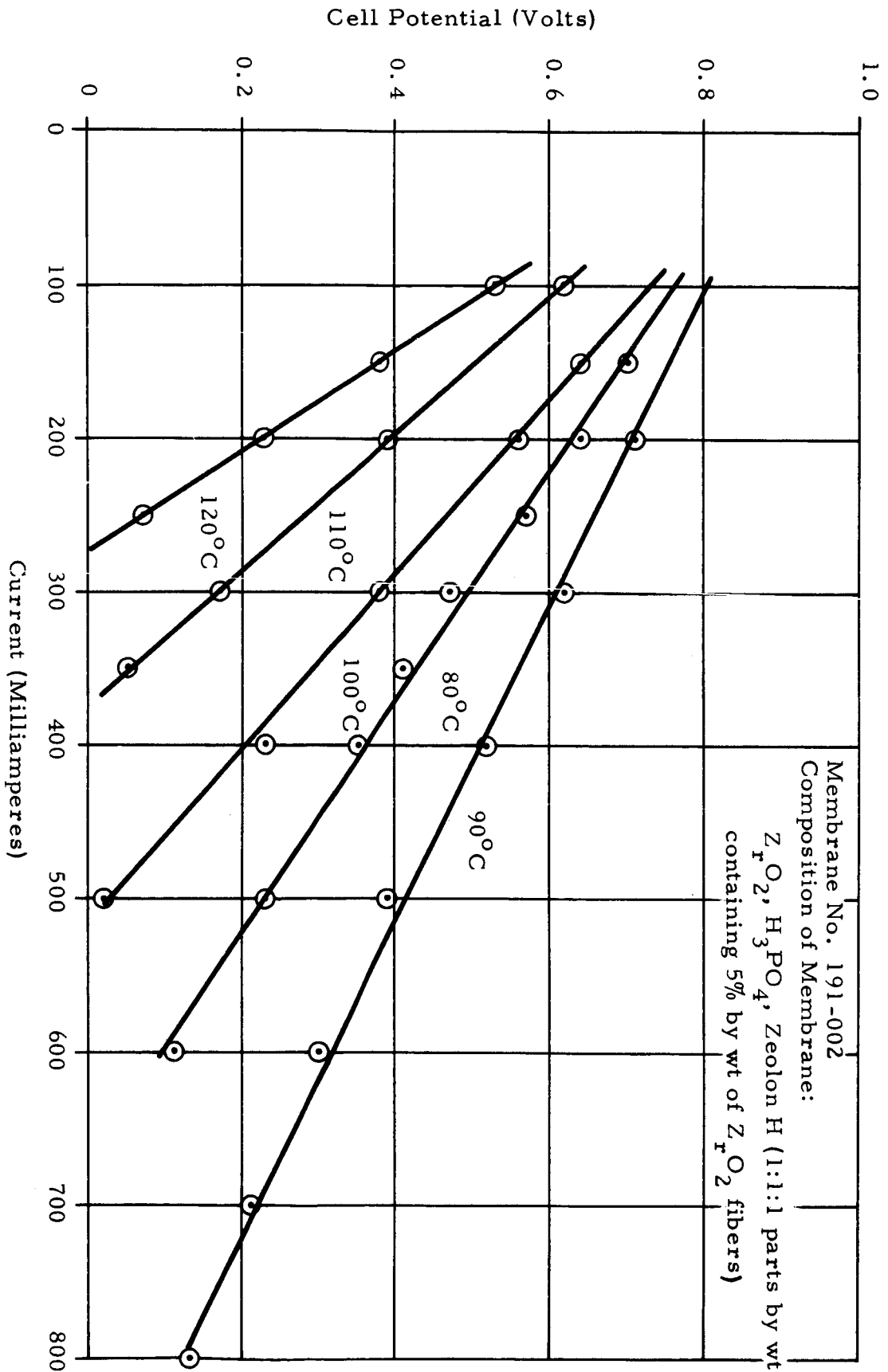


Figure 7. Influence of Temperature upon Characteristics of a Hydrogen-Oxygen Membrane Fuel Cell